June 10th, 2024

Joint paper on "a new computational method for a large-scale and high-accuracy quantum chemistry calculation on quantum computers" published in a Nature Research Journal
～Development of novel quantum computation method combining tensor network and Quantum Monte Carlo～

The Mitsubishi Chemical Group※1(hereafter, "Mitsubishi Chemical"), Keio University (headquartered in Minato, Tokyo, President Kohei Itoh) and IBM Japan, Ltd. (hereafter, "IBM", headquartered in Minato, Tokyo, General Manager Akio Yamaguchi), are pleased to announce the publication of a paper describing the research results of a new computational method for a large-scale and high-accuracy quantum chemistry calculation on quantum computers, a joint project from IBM Quantum Network Hub at Keio University Quantum Computing Center※2, in *npj Quantum Information*※3, a world-renowned Nature Research Journal.

Mitsubishi Chemical, Keio University and IBM Japan developed approaches to accurately calculate quantum mechanical ground states of large chemical systems, and the approaches are called HTN+QMC method and pseudo-Hadamard test technique; HTN+QMC combines a partitioning method called Hybrid Tensor Network (HTN), with a high-accuracy calculation method called Quantum Monte Carlo (QMC), to calculate the energy of large molecules and solids. The pseudo-Hadamard test technique efficiently calculates the overlap between quantum states from quantum circuits. By applying these methods to quantum chemistry calculations for photochromic model molecules on the gate-based quantum computer IBM Quantum System One, ground state energies were successfully computed as 0.042 ± 2.0 milli-Hartree, an accuracy comparable to that of a noiseless simulator.

This research is expected to pave the way for accurate analysis of the physical properties of large-scale molecules and solids, which are beyond the size that can be handled by a stand-alone quantum computer.

We plan to continue research in the area of using quantum computers to accelerate the development of a wide range of new materials.

※1：The Mitsubishi Chemical Group is the collective name for Mitsubishi Chemical Group Corporation and its group companies.

※2：Opened in May 2018 by Keio University and IBM Japan on the Yagami Campus of the university's Faculty of Science and Technology, the IBM Quantum Network Hub at Keio University brings together academia and industry with Mitsubishi Chemical as founding member. It is also the first IBM Quantum Hub in Asia to leverage IBM Quantum Systems, the most advanced quantum computers developed by IBM and delivered via the cloud.

※3：The published paper URL＝https://www.nature.com/articles/s41534-024-00851-8

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Reference

【Key research findings】

- A method for a large-scale and high accuracy quantum chemistry calculation called HTN+QMC※4 was developed by combining a partitioning method, Hybrid Tensor Network, and a high accuracy calculation method, Quantum Monte Carlo.
- A pseudo-Hadamard test technique was developed to efficiently calculate the overlap between quantum states.
- We expect the method to play an important role in accurately understanding the physical properties of large molecules and solids.

【Background】

The physical properties of molecules and solids can be determined by calculating stationary states in the material. However, since the cost of calculating stationary state increases exponentially with the number of electrons, calculations are currently performed using approximations. DFT ※5, which incorporate knowledge of electronic correlation into approximations, is widely used in the calculation of the ground state※6 of electrons, but in principle cannot accurately calculate the electronic structure of strongly correlated systems such as materials in the presence of strong Coulomb repulsion.

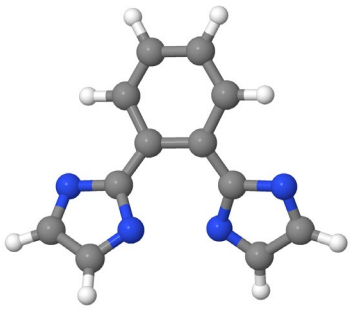

Fig. 1. Structure of MonoArylBiImidazole. The gray, white, and blue elements represent a carbon, hydrogen, and nitrogen atoms, respectively.

Quantum computers are gaining interest as a potential solution to this issue because they can perform calculations that cannot be performed by conventional (classical) computers by leveraging quantum entanglement※7 and quantum superposition※8. However, current quantum computers are limited in the number of qubits and the number of gates available, so calculating physical properties from quantum first principles will require a large-scale, high-accuracy calculation method that can exceed the performance of a quantum device alone.

【Achievements of this research project】

In this study we developed: [A] HTN+QMC, which combines a partitioning method and a high-accuracy calculation method, and [B] the pseudo-Hadamard test technique, which efficiently calculates the overlap between quantum states necessary for HTN+QMC.

[A] Development of HTN+QMC, a large-scale high-accuracy energy calculation method based on the combination of the partitioning method and high accuracy calculation methods.

In this study, a hybrid tensor network (HTN) was employed as the partitioning method and quantum Monte Carlo (QMC) was used as the high-accuracy calculation method.

The hybrid tensor network [Figure 2(a)] method is a hybrid quantum-classical method that divides a quantum state that is larger than a single quantum computer into smaller tensors (blocks). In our case, each tensor is handled by a quantum computer, and the connections between tensors are handled by a classical computer. Since the method uses both quantum and classical computers, it is called the hybrid tensor network method. This design allows us to perform large calculations while maximizing the use of the quantum computer. For example, by using HTNs, a 100-qubit quantum computer can be used to generate a quantum state of 10,000-qubit system in principle.

Quantum Monte Carlo [Figure 2(b)] is a high-accuracy energy calculation method. The use of a quantum computer in quantum Monte Carlo calculations can improve accuracy. For example, the energy calculation accuracy is expected to be improved by using quantum states generated by

quantum circuits※9.

The HTN+QMC [Figure 2(c)] method we developed uses an HTN to generate quantum states in order to perform quantum Monte Carlo on a chemical system with spin orbitals larger than the size of the quantum computer.

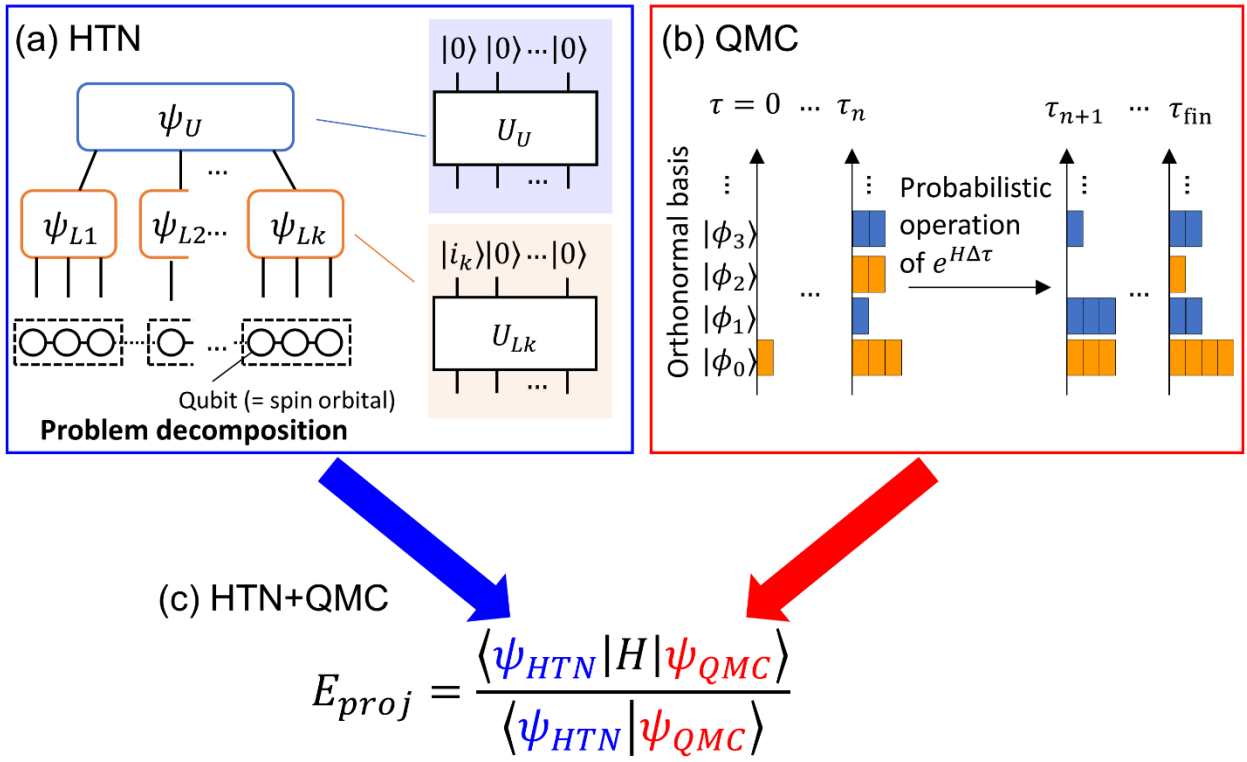

Fig. 2.

(a) Hybrid Tensor Network (HTN). In this study, we adopted a two-layer tree-type tensor network consisting of lower tensors (orange) and an upper tensor (blue). Each tensor consists of a quantum circuit. The model is partitioned into groups of qubits (= spin orbitals), and each of the partitioned models is assigned to each lower tensor. The upper tensor is used to integrate the results of the lower tensor.

(b) Quantum Monte Carlo (QMC). We adopted full configuration interaction Monte Carlo, and the amplitude of the quantum state is evolved based on a stochastic imaginary-time evolution to generate an approximate ground state. The color of the amplitude indicates the sign.

(c) HTN+QMC. The accuracy of QMC result for large systems can be improved by using the quantum states generated by HTN in the calculation of the projected energy E_{proj}, i.e., an energy estimator.

[B] Development of the pseudo-Hadamard test, a technique to efficiently calculate the overlap between quantum states

Calculating the overlap between quantum states is necessary to perform HTN+QMC.As shown in the upper panel of Figure 3(a), the calculation steps are as follows: [step A] prepare quantum gates to generate approximate ground states by optimization, and [step B] obtain the overlap by entangling the obtained gates and ancilla qubits, or more precisely, by performing the controlled operation (red part of the figure). Step B is called the Hadamard test, and the computational cost of the test increases with the number of the prepared gates. This is a particularly difficult issue for superconducting quantum computers, as computational cost is high for distant qubit operations. Using our pseudo-Hadamard test [Figure 3(a), bottom] the cost increase in the calculation of overlapping states shown in step B can be avoided by involving an ancilla qubit in the quantum gate optimization, shown in step A [Figure 3(b)]. In a conventional overlap calculation using the Hadamard test, controlled gate operations must be performed; in contrast, our method calculates the overlap without explicitly performing controlled gate operations. Thus, we name the method the pseudo-Hadamard test.

Using a combination of the approaches we developed [A] and [B], we performed ground-state

calculations for the photochromic※10 model molecule MonoArylBiImidazole (Figure 1) on the gate-based quantum computer IBM Quantum System One. We calculated the ground state as 0.042± 2.0 milli-Hartree, which is an accuracy comparable to a noiseless simulator. The accuracy typically required to understand chemistry phenomena is 1.6 milli-Hartree.

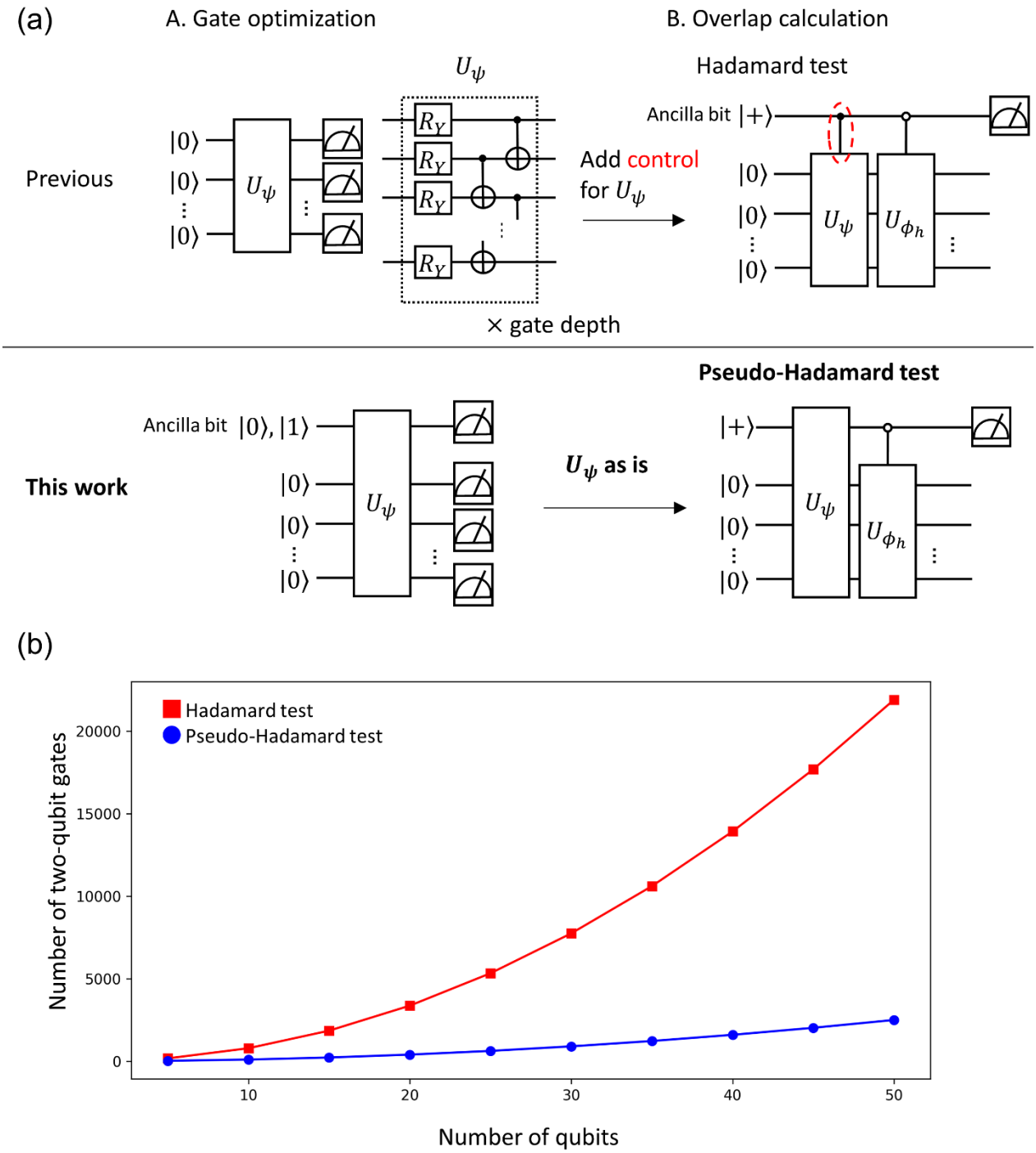

Fig 3.

(a) Overview of the pseudo-Hadamard test. We calculate the overlap $\langle\psi|\phi_h\rangle$ between the quantum state that we generate in a quantum circuit $|\psi\rangle$ and the orthogonal basis of index $h, |\phi_h\rangle$ [see Figure 2(a)]. The upper and lower panels show the conventional and developed techniques, respectively. Here, U_ψ is a gate to generate $|\psi\rangle$, which may consist of a group of gates as shown in the center of upper panel. Gates within the dotted lines are repeated multiple (depth) times. Note that U_{ϕ_h} is a gate to generate $|\phi_h\rangle$ and can be easily implemented.

(b) Comparison of the number of two-qubit gates※11 required for the Hadamard test and pseudo-Hadamard test.

【Future work】

Although this HTN+QMC method was developed to calculate the ground states of chemical

systems, it can be applied to a wide range of tasks such as machine learning and optimization. The pseudo-Hadamard test is also not limited to HTN+QMC, but can be applied to calculation of overlapping states and transition amplitude between any quantum states. This research paves the way to highly accurate calculations using quantum computers, not only for chemistry calculations, but also for large-scale calculation tasks.

【Glossary】

※4：HTN+QMC
Abbreviation for Hybrid Tensor Network + Quantum Monte Carlo which is developed in this study.

※5：DFT
Abbreviation for density functional theory.

※6：Ground state
The lowest energy electronic state.

※7：Quantum entanglement
The property of a qubit to take on multiple states simultaneously.

※8：Quantum superposition
A relationship in which states are mutually dependent among multiple qubits.

※9：Quantum circuit
Combines quantum gates and measurements that control a quantum state of qubits.

※10：Photochromic
A property of a material that changes color in response to changes in light intensity.

※11：Comparison for the numbers of two-qubit gates
We assume a nearest neighbor connected device and that the gate depth is equal to the number of system qubits. In the Hadamard test, for each CNOT gate, we estimated the case where a round-trip using qubit swap occurs from the original position to the adjacent auxiliary qubit when performing the control operations from the auxiliary qubit.

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【Press Release References】

・ A joint paper on “a new computational method for energy calculations of photonic materials on quantum computers” was published in a Nature Research Journal（Feb 9, 2023）

・ A Joint Paper on Prediction of Optical Properties of OLED Materials was Published in a Nature Research Journal（May 26, 2021）